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Far-infrared electromagnetic radiation is emitted by coherent plasmon oscillations in bulk (111) InSb. The oscillations are excited by near-infrared ultrashort laser pulses and characterized as a function of temperature. The coherent plasmon frequency is determined by the sum of the intrinsic electron concentration and donor doping density. The amplitude of the oscillations decreases with increasing temperature due to a weakening of the photo-Dember starting mechanism.

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Coherent plasmons in InSb

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Abstract

Far-infrared electromagnetic radiation is emitted by coherent plasmon oscillations in bulk (111) InSb. The oscillations are excited by near-infrared ultrashort laser pulses and characterized as a function of temperature. The coherent plasmon frequency is determined by the sum of the intrinsic electron concentration and donor doping density. The amplitude of the oscillations decreases with increasing temperature due to a weakening of the photo-Dember starting mechanism.

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Semiconductors can produce far-infrared electromagnetic transients when illuminated with ultrashort laser pulses. Applications that use these THz frequency pulses are now being explored, examples of which are three-dimensional imaging and time-resolved spectroscopy [1]. It has been established that THz emission from bulk semiconductors originates from distinct phenomena such as nonlinear optical rectification of the ultrashort laser pulse at high irradiance and ultrafast transport of charges at low irradiance. Both mechanisms have been identified in experiments with bulk InSb, the latter effect displaying a strong dependence on lattice temperature and wavelength of the pump laser pulses [2–8]. A physical picture of a current surge by photo-excited electron-hole pairs has emerged, in which the mobility of the carriers plays a fundamental role [9, 10]. Experimental characterization of THz pulses has been hampered, however, by the limited bandwidth of gated photoconducting detection antennas and the strong dispersion near the TO phonon frequency of crystals used in time-resolved electro-optic sampling [11].

Experiments with other bulk semiconductors have revealed THz radiation originating from the coherent motion of bulk plasmons [12] and phonons [13]. Using an antenna with response up to 7 THz, Gu *et al.* detected coherent plasmons and phonons in the radiation spectra of InSb [14]. They found that the plasma frequency was independent of excitation power and identified the presence of an ultrafast, ambipolar diffusion of photocarriers near the sample surface (i.e., a photo-Dember current). Their experiments were performed at room temperature where the coherent oscillation signals are very weak. In this Letter, we report results of a temperature-dependent study of THz emission from bulk (111) InSb. We find that the emitted radiation is dominated by coherent plasmon oscillations of the background electron gas, *not* a current surge of photocarriers. The temperature dispersion of the plasma frequency agrees quantitatively with a model that uses no fitting parameters.

We measure the coherence of the THz pulses using a modified Michelson interferometer arrangement and a composite Si bolometer cooled with liquid helium [12]. This technique provides a uniform, broadband response in the spectral region of interest, although it cannot determine the phase of the radiated field relative to the excitation pulse. Near-infrared pump pulses are obtained from a mode-locked Ti:sapphire laser with duration of 30-fs and center wavelength $\sim$ 760 nm. Free-standing InSb crystal substrates (thickness: 1 mm; $E_g = 0.228$ eV at 80 K) are grown in the (111) orientation and lightly n-doped with Te. Samples are attached to the coldfinger of an optical cryostat and irradiated with p-polarized laser light

at an incidence angle of 45°. The pump light is focused on the samples with an irradiance $\sim 50\%$ below the level at which broadband optical rectification can be observed. The THz pulses co-propagate with the reflected laser beam, which is attenuated with a $\sim 50\ \mu m$ teflon film to prevent it from reaching the bolometer detector. The THz beam path is purged with dry nitrogen gas to mitigate water vapor absorption. A spatial filter reduces the incoherent background signal due to bandgap recombination radiation in InSb. The MgF_2 cryostat window limits the maximum far-infrared detection frequency to ~ 3 THz.

Time-resolved interference traces depicting radiation from coherent plasmon oscillations are presented in Fig. 1. The sample has a donor concentration of $N_D = 3 \times 10^{14}\ cm^{-3}$. With increasing temperature we observe: i) a blue-shift of the plasma frequency, ii) a decrease of the dephasing time from 1.7 ± 0.2 ps at 80 K to 0.7 ± 0.1 ps at 200 K, and iii) a pronounced decrease in the amplitude of the oscillations.

Coherent plasmons are generated by the motion of the cold electrons; THz radiation from the inhomogeneous distribution of optically injected electron-hole pairs (estimated density $\sim 10^{19}\ cm^{-3}$) is not detected in our experiment. The background electron concentration is the sum of the donor density plus intrinsic thermal excitations across the bandgap. Shallow level impurities do not freeze-out in InSb, so the temperature-dependent electron concentration is [15]:

$$N = N_D + 5.7 \times 10^{14}\ T^{3/2}\ \exp(-0.125/kT) \tag{1}$$

where N is in units of cm^{-3} and T is in Kelvin. The electron plasma frequency is (cgs):

$$\nu_p = \sqrt{\frac{Ne^2}{\pi \varepsilon_\infty m_{opt}}} \tag{2}$$

where $\varepsilon_\infty = 15.64$ and e is the electronic charge. The optical effective mass of the electrons (m_{opt}) changes with both temperature and electron density due to the nonparabolic conduction band of InSb [16]. The donor ions and holes are both assumed to have negligible motion (i.e., infinite mass) and do not affect the calculation. Fig. 2 shows that the coherent plasmon frequencies obtained from Fourier transforming the signals in Fig. 1 are well described by this model. For $T > 150$ K, the intrinsic carrier concentration is larger than the donor density and the oscillation frequency strongly blue-shifts. For comparison, the measured coherent plasmon frequencies for a sample doped with $N_D = 1.1 \times 10^{15}\ cm^{-3}$ are shown. Below 200 K, intrinsic contributions to the electron concentration are negligible

and the oscillation frequency is determined by the constant donor density ($\nu_p = 0.5 \pm 0.05$ THz).

The decay of cold plasmon oscillations is greatly influenced by the mobility of the bulk semiconductor [17]. In the temperature range of our experiments, electron mobility is primarily due to scattering with LO phonons. Hall mobility measurements of the sample in Fig. 1 indicate an electron momentum relaxation time of 3.2 ps at 77 K, decreasing to ~ 1.5 ps at 200 K. Although temperature scaling of LO phonon scattering follows the trend of our data, the measured dephasing times are 2 times shorter than indicated by the mobility of the unexcited sample. This suggests that coherent plasmon scattering with optically excited holes also contributes to the dephasing rate [18, 19].

The photo-Dember effect is the starting mechanism for coherent plasmon oscillations [14]. The strong reduction in the coherent plasmon amplitude with increasing temperature in Fig. 1 is explained by the decreased mobility of the hot photocarriers. The rate of carrier scattering with LO phonons increases with lattice temperature and damps out the diffusion current transient. Fig. 3 displays the THz radiation signal obtained at 300 K; the cryostat window is removed to extend the high frequency response past 10 THz. The coherent plasmon is extremely weak while pronounced coherent LO phonon oscillations are visible at 6 THz (Fourier transform spectrum shown in the inset). We have previously shown that when ultrafast polarization changes are minimized, coherent oscillations can be excited by impulsive stimulated Raman scattering (ISRS) and radiate into free space [20]. At 300 K (Fig. 3), the photo-Dember current has been sufficiently weakened to allow the observation of narrow bandwidth radiation from coherent phonons driven by ISRS.

In summary, we studied coherent plasmon oscillations in bulk (111) InSb by characterizing the far-infrared radiation they emit. The plasma frequency can be adjusted by thermal excitations and doping. With increasing temperature, the source of the THz radiation transitions from coherent plasmon emission started by the an ultrafast polarization change resulting from the photo-Dember effect to coherent LO phonon oscillations excited in ISRS.

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[1] B. Ferguson and X-C Zhang, Nature Materials **1**, 26 (2002).

[2] B.B. Hu, X-C Zhang, and D.H. Auston, Appl. Phys. Lett. **57**, 2629 (1990).

[3] S.C. Howells, S.D. Hererra, and L.A. Schlie, Appl. Phys. Lett. **65**, 2946 (1994).

[4] S.C. Howells and L.A. Schlie, Appl. Phys. Lett. **67**, 3688 (1995).

[5] R. McLaughlin, Q. Chen, A. Corchia, C.M. Cisela, D.D. Arnone, X-C. Zhang, G.A.C. Jones, E.H. Linfield, and M. Pepper, J. Mod. Opt. **47**, 1847 (2000).

[6] S. Kono, P. Gu, M. Tani, and K. Sakai, Appl. Phys. B **71**, 901 (2000).

[7] P. Gu, M. Tani, S. Kono, K. Sakai, and X-C Zhang, J. Appl. Phys. **91**, 5533 (2002).

[8] H. Takahashi, Y. Suzuki, M. Sakai, S. Ono, N. Sarakura, T. Sugiura, T. Hirosumi, and M. Yoshida, Appl. Phys. Lett. **82**, 2005 (2003).

[9] X-C Zhang and D.H. Auston, J. Appl. Phys. **71**, 326 (1992).

[10] M.B. Johnston, D.M. Whittaker, A. Corchia, A.G. Davies, and E.H. Linfield, Phys. Rev B **65**, 165301 (2002).

[11] A. Leitenstorfer, S. Hunsche, J. Shah, M.C. Nuss, and W.H. Knox, Appl. Phys. Lett. **74**, 1516 (1999).

[12] R. Kersting, K. Unterrainer, G. Strasser, H.F. Kauffman, and E. Gornik, Phys. Rev. Lett. **79**, 3038 (1997).

[13] T. Dekorsy, H. Auer, C. Waschke, H.J. Bakker, H.G. Roskos, H. Kurz, V. Wagner, and P. Grosse, Phys. Rev. Lett. **74**, 738 (1995).

[14] P. Gu, M. Tani, K. Sakai, and T-R Yang, Appl. Phys. Lett. **77**, 1798 (2000).

[15] E.H. Putley, Proc. Phys. Soc. **73**, 280 (1959).

[16] M.P. Hasselbeck and P.M. Enders, Phys. Rev. B **57**, 9674 (1997).

[17] R. Kersting, J.N. Heyman, G. Strasser, and K. Unterrainer, Phys. Rev. B **58**, 4553 (1998).

[18] M. Hase, S. Nakashima, K. Mizoguchi, H. Harima, and K. Sakai, Phys. Rev. B **60**, 16526 (1999).

[19] M.P. Hasselbeck, D. Stalnaker, L.A. Schlie, T.J. Rotter, A. Stintz, and M. Sheik-Bahae, Phys. Rev. B **65**, 233203 (2002).

[20] M.P. Hasselbeck, L.A. Schlie, and D. Stalnaker, Appl. Phys. Lett., in press (2004).

Figure Captions

Fig. 1. Temperature-dependent THz interferograms of (111) InSb showing bulk plasmon oscillations. Curves are plotted on the same relative scale and displaced vertically for clarity.

Fig. 2. Coherent plasmon frequency obtained for two samples with different donor densities. The frequency increases (filled circles) when the intrinsic carrier concentration becomes comparable to the donor density as predicted by the model (solid line). When the doping density exceeds the intrinsic density throughout the experimental temperature range (triangles), the plasma frequency does not change.

Fig. 3. Autocorrelation signal for the sample in Fig. 1 at room temperature. The cryostat window is removed to extend the frequency response. Fourier transform of this signal (inset) shows the emitted radiation is dominated by coherent LO phonon oscillations at 6 THz.

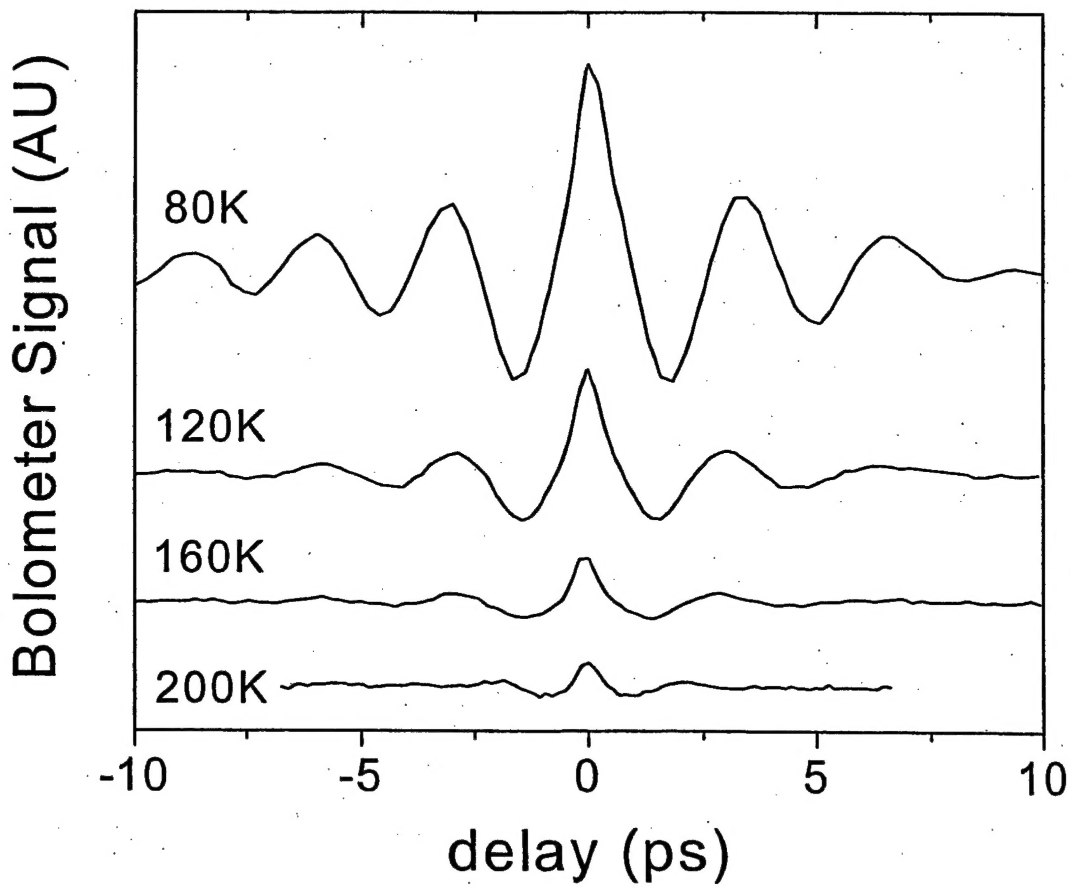

Fig. 1, Hasselbeck et al

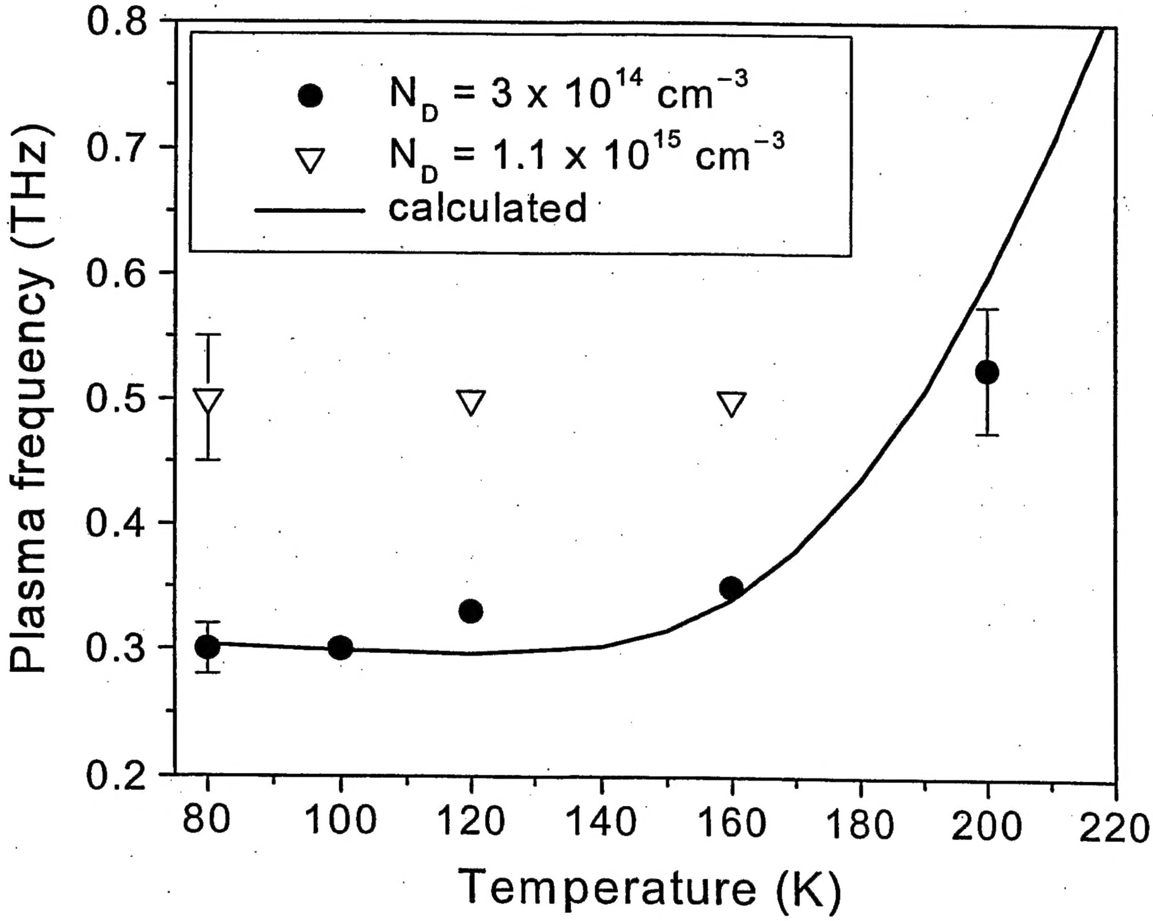

Fig. 2, Hasselbeck et al

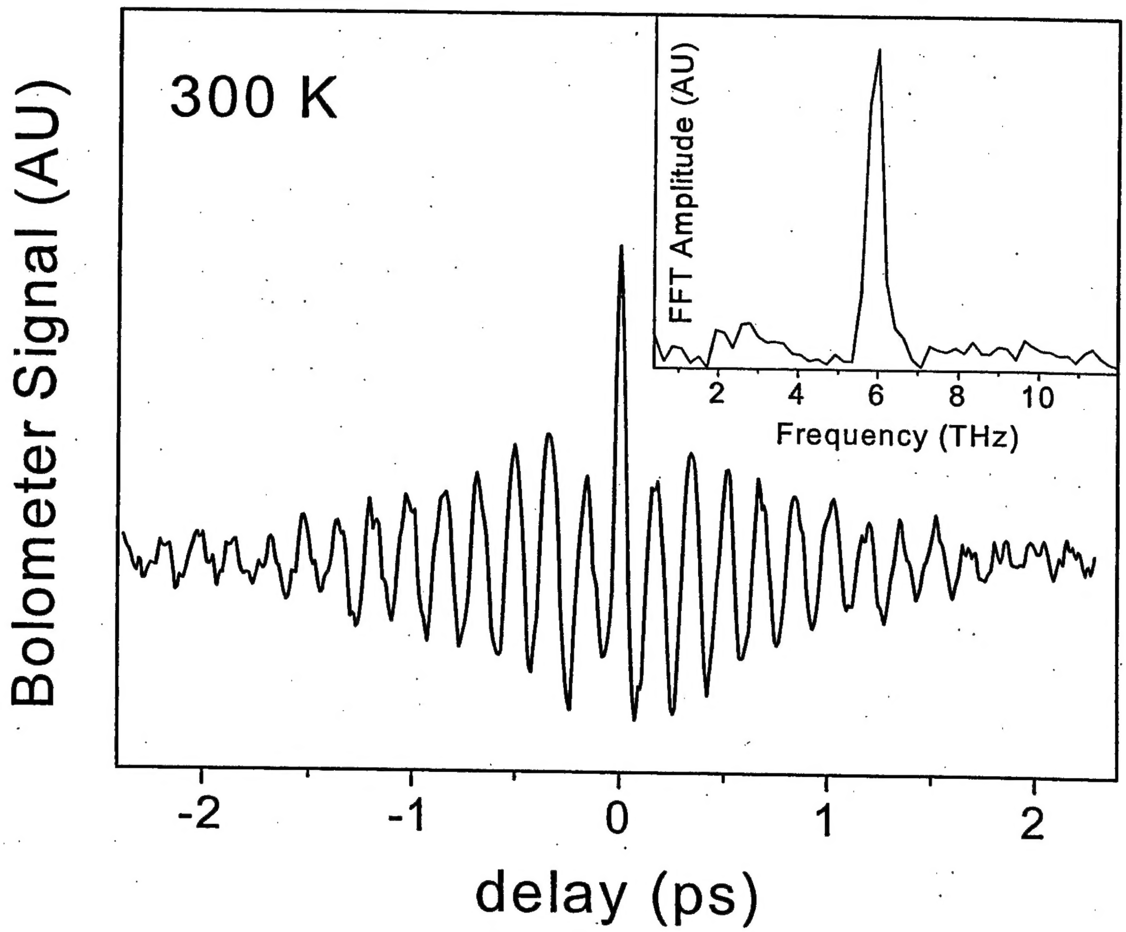

Fig. 3, Hasselbeck et al